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Demystifying mysterious time and space evolution in state-of-the art graphite-silicon composite battery electrodes

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The reaction processes in Li-ion batteries can be highly heterogeneous at the electrode scale, leading to local deviations in the lithium content or local degradation phenomena. To access the distribution of lithiated phases throughout a high energy density silicon-graphite composite anode, we apply correlative operando small- and wide-angle X-ray scattering (SAXS and WAXS) tomography.

In-plane and out-of-plane inhomogeneities are resolved during cycling at moderate rates, as well as during relaxation steps performed at open circuit voltage (OCV) at given states of charge. Lithium concentration gradients in the silicon phase are formed during cycling, with regions close to the current collector being less lithiated when charging.

In relaxing conditions, the multi-phase and multi-scale heterogeneities vanish to equilibrate the chemical potential. In particular, Li-poor silicon regions pump lithium ions from both lithiated graphite and Li-rich silicon regions.

This charge redistribution between active materials is governed by distinct potential homogenization throughout the electrode and hysteretic behaviours. Such intrinsic concentration gradients and out-of-equilibrium charge dynamics, which depend on electrode and cell state of charge, must be considered to model the durability of high capacity Li-ion batteries.

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